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Microwave coherent control of ultracold ground-state molecules formed by short-range photoassociation

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We report the observation of microwave coherent control of rotational states of ultracold $^{85}\text{Rb}^{133}\text{Cs}$ molecules formed in their vibronic ground state by short-range photoassociation. Molecules are formed in the single rotational state $X(v=0, J=1)$ by exciting pairs of atoms to the short-range state $(2)^3\Pi_0-(v=11, J=0)$, followed by spontaneous decay. We use depletion spectroscopy to record the dynamic evolution of the population distribution and observe clear Rabi oscillations while irradiating on a microwave transition between coupled neighbouring rotational levels. A density-matrix formalism that accounts for longitudinal and transverse decay times reproduces both the dynamic evolution during the coherent process and the equilibrium population. The coherent control reported here is valuable both for investigating coherent quantum effects and for applications of cold polar molecules produced by continuous short-range photoassociation.

Introduction

Recent decades have witnessed fast developments in the study of ultracold molecules, which are of great interest in both physics and chemistry^{1–5}. Ultracold polar molecules have abundant internal states and interact via strong, anisotropic, and long-ranged dipolar interactions. They have potential applications in ultracold chemistry^{6–10}, precision measurement^{11,12}, quantum simulation¹³ and quantum computation^{14,15}.

All these applications require cold polar molecules in a well-defined initial state. Such molecules may be produced in a variety of ways. There have been rapid recent developments in direct laser cooling and magneto-optical trapping (MOT) to produce ultracold polar molecules in a single quantum state^{16–18}. However, this technique is limited to a small class of molecules with nearly closed laser-cooling transitions, and currently produces only very low phase-space densities. An alternative approach is to form molecules from pairs of ultracold atoms by magnetoassociation using a magnetic field ramp. Such molecules are

initially in weakly bound Feshbach states of the electronic ground state, but it is often possible to transfer them coherently to the vibronic ground state by stimulated Raman adiabatic passage (STIRAP)¹⁹. In favourable cases, this can produce substantial densities of molecules in a single hyperfine and Zeeman state^{20–25}. A further class of methods is based on photoassociation (PA) using laser beams. One-photon PA associates pairs of atoms to form cold molecules in an excited electronic state, usually highly vibrationally excited. These molecules soon decay spontaneously to the ground excited state; they are usually still in highly excited vibrational states, but may be transferred to the vibronic ground state either by a pump-dump scheme²⁶ or by STIRAP²⁷.

Coherent methods can form molecules only once per experimental cycle. By contrast, methods that involve spontaneous emission allow accumulation of molecular density. In particular, short-range photoassociation offers a simple optical pathway from atoms to molecules; it allows accumulation of cold molecules in the vibronic ground state, and can be applied to systems that are not amenable to magnetoassociation. Short-range PA has been implemented for various heteronuclear molecules^{28–32}. We have recently formed polar $^{85}\text{RbCs}$ molecules in this way, trapped them optically, and measured atom-molecule collision rates³³, while Passagem *et al.* have developed a special laser that can simultaneously photoassociate ^{85}Rb atoms and trapping the resulting Rb_2 homonuclear molecules³⁴.

Once molecules have been produced in a single quantum state, coherent control is needed. Polar molecules have al-

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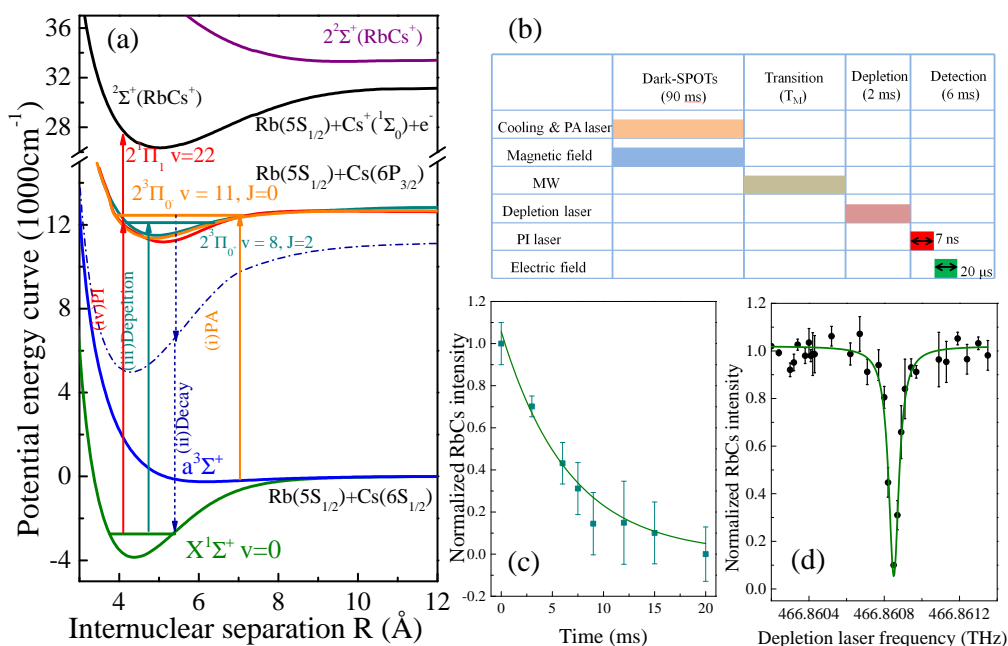


Fig. 1 (Color online) Experimental overview. (a) Optical pathways to produce and detect ultracold $^{85}\text{Rb}^{133}\text{Cs}$ molecules in the state $X^1\Sigma^+(v=0)$. (b) Time sequence. (c) Lifetime measurement of formed molecules in the absence of both the depletion laser and the MW field. (d) Depletion spectroscopy measurement in the presence of only depletion laser.

lowed microwave (MW) transitions between neighbouring rotational states³⁵, which may be driven with very high resolution. Coherent control using such transitions has been achieved both for molecules produced by STIRAP, including $^{40}\text{K}^{87}\text{Rb}$ ³⁶, $^{23}\text{Na}^{40}\text{K}$ ^{37,38}, $^{87}\text{Rb}^{133}\text{Cs}$ ³⁹, $^{23}\text{Na}^{87}\text{Rb}$ ⁴⁰, and for CaF produced by direct laser cooling⁴¹. Such control is at the heart of nearly all proposals for applications, such as simulating quantum magnetism^{42,43}, coupling quantum qubits^{44,45}, controlling state-dependent chemical reactions⁴⁶, inducing dipolar interaction for topological phase⁴⁷, enhancing evaporative cooling⁴⁸ and synthetic dimensions⁴⁹.

Here we report coherent control of rotational states of ultracold polar molecules produced in the lowest vibronic state by continuous photoassociation. We observe clear Rabi oscillations between neighbouring rotational states. We use a density-matrix formalism that accounts for longitudinal and transverse decay times to analyze the evolution of the population distributions, determine the coherence time, and understand the equilibrium state.

Experimental setup

A full description of our apparatus has been given previously⁵⁰. We therefore give only a summary of the main procedures and parameters here; further details can be found in ref.⁵⁰.

The precooled atom samples are prepared as before, but here we photoassociate via an excited molecular state that decays to a particularly simple rotational distribution in the lowest vibronic state. This facilitates subsequent control of the quantum state. We

also use a different time sequence, which is shown in Fig. 1(b). This separates the microwave coupling step from population measurement by depletion spectroscopy, allowing us to investigate the coherence of the microwave transition.

Under a vacuum background pressure around 3×10^{-7} Pa and at a magnetic gradient around 15 G/cm, we trap a mixed atomic cloud that consists of 1×10^7 Rb atoms in the state $5S_{1/2}$ ($F=2$) and 2×10^7 Cs atoms in the state $6S_{1/2}$ ($F=3$). The number densities of Rb and Cs atoms are $8 \times 10^{10} \text{ cm}^{-3}$ and $1 \times 10^{11} \text{ cm}^{-3}$, respectively. The translational temperature of the mixture is measured by time-of-flight imaging to be around 100 μK .

We carry out photoassociation using the optical pathways shown in Fig. 1(a), with potential energy curves based on the results of Refs.⁵¹ and⁵². The chosen intermediate molecular state is $2^2\Pi_0$ ($v=11, J=0$). As shown by Shimasaki *et al.*³¹, this state decays to $X^1\Sigma^+(v=0)$ by two-photon cascade. The molecules formed are then ionized by a tunable pulsed dye laser, accelerated by a pulsed electric field, and finally detected by a pair of micro-channel plates. In the absence of both the depletion laser and the MW field, the lifetime is measured to be 6.6(6) ms, as shown in Fig. 1(c). When parity is conserved, the two-photon decay produces only one rotational state, $J=1^-$, although Ref.³¹ observed small populations in $J=0^+$ and 2^+ as well, due to Stark mixing induced by the residual static electric field in their experiment.

Figure 1 (d) shows the result of depletion spectroscopy for the molecules we produce in the absence of the MW field. The in-

interaction time and intensity of the depletion laser are 2 ms and 1 mW/mm² respectively. The single transition observed may be assigned as $X^1\Sigma^+(v=0, J=1)$ to $2^3\Pi_{0+}(v=8, J=2)$, based on the assignments in Fig. 2(b) of Ref.⁵⁰. The fractional depletion is greater than 90% and the transition from $J=3$ to 4 is not observed. This demonstrates that only the $J=1$ state of $X^1\Sigma^+(v=0)$ is significantly populated in the present experiment. Transitions originating in $J=0$ and 2 are also not observed, confirming that the influence of Stark mixing is not important here. The full width at half maximum (FWHM), obtained by fitting to a Lorentz lineshape, is found to be 73(5) MHz and is attributed to power broadening due to the depletion laser.

Experimental results and analysis

After production of molecules in the state $X^1\Sigma^+(v=0, J=1)$, we irradiate them with microwave radiation close to the $J=1 \rightarrow 2$ transition. The MW field is produced with a homemade coil and has no well-determined quantization axis. The resulting rotational population distributions are shown as a function of MW frequency in Fig. 2(a). The interaction time is chosen to be 2 ms, which is longer than the coherence time. The microwave intensity is around 1 μ W, which is enough lower than the saturation power to ensure sufficient signal-to-noise. We measure the radiant power using a microwave power meter (NRP-Z51, R&S) with a circle probe of diameter 2.3 cm. Since the power at the atomic cloud cannot be measured directly, we use the measured value at an equivalent distance from the homemade radiant coil. To measure the population in $J=1$, the frequency of the depletion laser shown in Fig. 1 is locked at the transition between $X^1\Sigma^+(v=0, J=1)$ and $2^3\Pi_{0+}(v=8, J=2)$, while for $J=2$ it is locked at the corresponding transition between $J=2$ and $J=3$. The population is obtained from one minus the ratio of the intensity of RbCs ions in the presence of the depletion laser to that in its absence. Fitting to Lorentzian lineshapes gives a resonant microwave frequency $\nu_{12} = 1988.62(1)$ MHz and FWHM $\gamma = 0.20(4)$ MHz from the population in $J=1$ and $\nu_{12} = 1988.60(1)$ MHz and $\gamma = 0.18(1)$ MHz from that in $J=2$. We can use either rotational state to probe coherent control. In the following, we focus on the population of the state $X^1\Sigma^+(v=0, J=1)$. Figure 2(b) shows the value of γ from the population in $J=1$ as a function of MW power. We use a simple model $\gamma = \gamma_0 \sqrt{1 + P/P_{\text{sat}}}$ ⁵³ to fit the experimental data, yielding $\gamma_0 = 0.20(8)$ MHz and $P_{\text{sat}} = 0.011(1)$ mW.

Figure 3 shows the population in $J=1$ as a function of MW irradiation time. The MW frequency is fixed at the central value fitted in Fig. 2(a). The measured MW power is 10 mW. The measured population shows a clear Rabi oscillation. We treat the two-level system theoretically using a density-matrix formalism under the electric-dipole and rotating-wave approximations. The time evolution is written as a pair of coupled equations⁵⁴,

$$\dot{\rho}_{21} = -(\Gamma_2 + i\Delta)\rho_{21} - i\frac{\Omega}{2}(\rho_{22} - \rho_{11}); \quad (1)$$

$$\dot{\rho}_{22} = \Gamma_1(\rho_{22}^0 - \rho_{22}) + i\frac{\Omega}{2}\rho_{12} - i\frac{\Omega}{2}\rho_{21}, \quad (2)$$

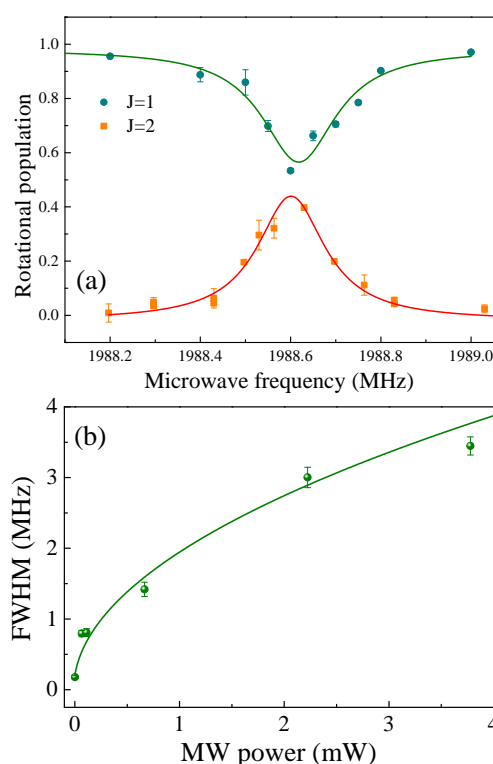


Fig. 2 (Color online) (a) The observed microwave transition in ultracold RbCs molecules, monitored with the depletion laser locked to the transition $2^3\Pi_{0+}(v=8, J=2) \leftarrow X^1\Sigma^+(v=0, J=1)$ (green squares) and the transition $2^3\Pi_{0+}(v=8, J=3) \leftarrow X^1\Sigma^+(v=0, J=2)$ (orange squares). The curves are fitted to a Lorentzian lineshape. (b) The FWHM width γ as a function of microwave power.

with parameters $\Gamma_1 = 1/T_1$ and $\Gamma_2 = 1/T_2$, where T_1 and T_2 are the longitudinal and transverse decay times. These times characterise the timescales for changes in population and for decoherence, respectively. Δ is the detuning of the applied fields and $\Omega = \mu_{12}E/\hbar$ is the Rabi frequency, where μ_{12} is the transition dipole moment (TDM) and E is the MW amplitude. The coupled differential equations are solved numerically, with the initial condition $\rho_{11}^0 = 1$ at $t = 0$ and the constraints $\rho_{11} + \rho_{22} = 1$ and $\rho_{12} = \rho_{21}^*$.

The curve in Fig. 3 shows the numerical simulation of ρ_{11} . In the simulation, the MW frequency is on resonance and the longitudinal decay time T_1 is chosen to be 6.6 ms, which is the measured lifetime of the molecules from Fig. 1(c). A satisfactory simulation is obtained with Rabi frequency $\Omega = 1.70$ MHz and coherence time $T_2 = 1.5 \mu$ s. This value of T_2 is more than two orders of magnitude lower than in other systems^{36–41}, but is still sufficient to allow several coherent manipulations and to investigate coherence effects. The factors limiting coherence arise mainly from the higher temperature of the precooled atoms in the present work, from inelastic molecular collisions⁵⁵ and from unresolved hyperfine structure.

The nuclei ^{85}Rb and ^{133}Cs have spins $I_{\text{Rb}} = 5/2$ and $I_{\text{Cs}} = 7/2$. The dominant hyperfine interactions in states with $J > 0$ are nuclear electric quadrupole interactions, with coupling constants $(eQq)_{\text{Rb}} = -1.661$ MHz and $(eQq)_{\text{Cs}} = 0.054$ MHz, obtained by Aldegunde and Hutson⁵⁶ from electronic structure calculations

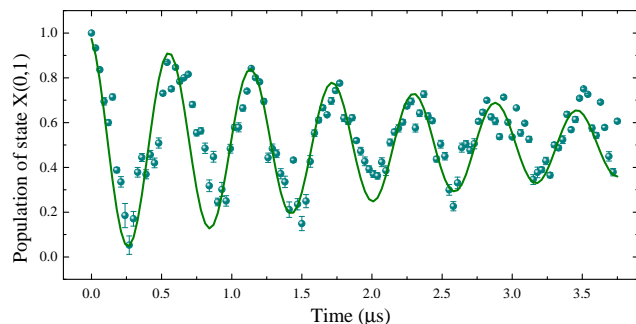


Fig. 3 (Color online) Evolution of the population of the initial state of the molecules, $J = 1$. The curve is a simulation based on Eq. 2. Each point represents the mean of 36 measurements.

using density functional theory. There are several other hyperfine terms, but their effect is at the kHz level. The allowed microwave lines for $J = 1 \rightarrow 2$ are expected to be spread over about 750 kHz; this is consistent with the observed coherence time $T_2 = 1.5 \mu\text{s}$ and may be compared with the observed FWHM $\gamma = 0.200(80)$ MHz.

The coherence time T_2 is independent of the experimental conditions, but the Rabi frequency Ω can be controlled. It depends externally on the MW amplitude and internally on the TDM. The MW amplitude E and power P are related to the intensity I by $I = \frac{1}{2} c n \epsilon_0 E^2 = P / \pi r^2$, so that $\Omega = (2 \mu_{12}^2 P / c n \epsilon_0 \pi r^2 \hbar^2)^{1/2}$. The Rabi frequency obtained from coherence measurements is shown as a function of MW power in Fig. 4(a). The fitted curve gives $\mu_{12} = 0.53(9)$ Debye. Here the error 0.09 Debye includes only the statistical error from the fit. There is an additional uncertainty in the TDM due to uncertainty in the MW power at the position of the atomic cloud, which we estimate as ± 0.03 Debye.

Our experiments use unpolarized microwaves, but in an isotropic environment the intensities may be obtained by considering plane-polarized light with the electric vector along a single direction⁵⁷. For z polarization, the TDM between the $M = 0$ components of $J = 1$ and 2 is $\mu_{12} = (4/15)^{1/2} \mu(v, J)$,⁵⁸ where $\mu(v, J)$ is the molecule-fixed dipole moment. This has been measured for $^{87}\text{RbCs}$ in its rovibronic ground state as $\mu(0, 0) = 1.225(11)$ Debye²². To obtain the corresponding value for $^{85}\text{RbCs}$, we solve the vibrational Schrödinger equation for each isotopolog using the ground-state RbCs interaction potential of Takekoshi *et al.*⁵⁹ and evaluate the expectation values $\mu(v, J)$ using the dipole-moment function of Fedorov *et al.*⁶⁰ The value obtained for $\mu(0, 0)$ in $^{85}\text{RbCs}$ is only about 9 parts in 10^7 smaller than for $^{87}\text{RbCs}$. The absolute value $\mu(0, 0) = 1.215$ D is less accurate than experiment, but the ratio between isotopologs is reliable. The dependence on rotational state is also negligible; it may be approximated by $\mu(v, J) = \mu(v, 0) + D_{\mu J}(J + 1)$, where $D_{\mu 0} = 2.34 \times 10^{-7}$ Debye for $^{85}\text{RbCs}$. The experimental value of $\mu(0, 0)$ ²² corresponds to $\mu_{12} = 0.633$ Debye, which is consistent with the present result in view of the uncertainty in the measured MW power.

At times much longer than the coherence time, the system reaches equilibrium and the population of the initial state $X^1\Sigma^+(v = 0, J = 1)$ becomes stable. Figure 4(b) shows the measured population of the state $X^1\Sigma^+(v = 0, J = 1)$ as a function of

MW power for an irradiation time of 2 ms, which is long enough for equilibrium to be established. It may be seen that the steady-state value is a little larger than 0.5. Ref.⁶¹ gives the steady-state population for the ideal resonant frequency. However, in a real experiment the detuning Δ is finite, though small, so here we use the generalized Rabi frequency $\tilde{\Omega} = \sqrt{\Omega^2 + \Delta^2}$ in place of the Rabi frequency,

$$\rho_{11}^{\text{eq}} = \frac{1}{2} \left[1 + \frac{\tilde{R}_3}{(1 + T_1 T_2 (\Omega^2 + \Delta^2))} \right]. \quad (3)$$

Here $\tilde{R}_3 = (1 - e^{-\hbar\omega_0/k_B T}) / (1 + e^{-\hbar\omega_0/k_B T})$ indicates the degree of mixedness of the reduced density matrix at temperature T in the absence of the external MW field and $\omega_0 = (E_2 - E_1)/\hbar$ is the resonant angular frequency. At the temperature of our experiment, $T = 100 \mu\text{K}$, \tilde{R}_3 is approximated as 1. The green dashed line and solid line in Fig. 4(b) show the simulated results when the detuning is zero (*i.e.* resonant) and 10 kHz (the minimum uncertainty in the FWHM of the MW spectra), respectively, using the measured lifetime $T_1 = 7$ ms and coherence time $T_2 = 4 \mu\text{s}$. Since there are large uncertainties in T_1 and T_2 and the product of them influences the steady-state population from Eq. 3, we have repeated the simulation for $\Delta = 10$ kHz with the experimental value of $T_1 T_2$ halved and doubled. Figure 4(b) shows that nearly all the measured populations are within the range of the simulated curves, which supports the theoretical model and estimates of uncertainty.

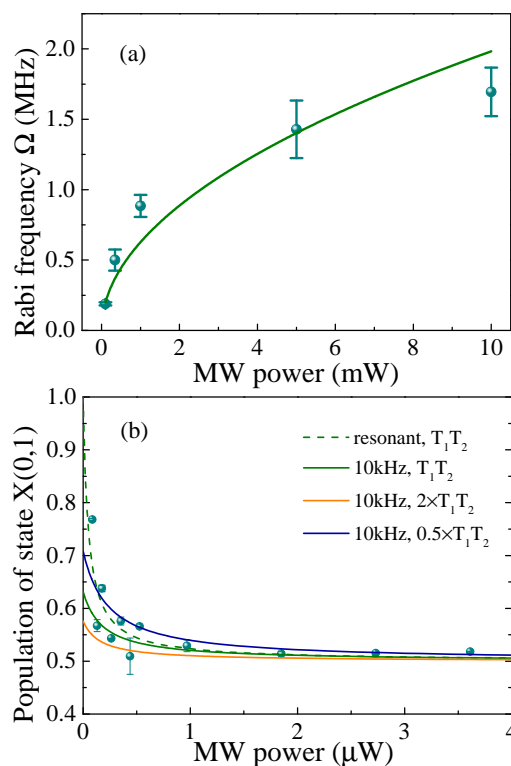


Fig. 4 (Color online) (a) Dependence of the Rabi frequency extracted from the coherence measurements on MW power. (b) The population of RbCs molecules in the initial state as a function of MW power, after irradiation for 2 ms.

Conclusion

In conclusion, we have demonstrated MW coherent control of ultracold polar $^{85}\text{RbCs}$ molecules formed by continuous short-range photoassociation from a cold atomic mixture. We observe clear Rabi oscillations and simulate them by adding decay terms to the classical Hamiltonian of a two-level system in a monochromatic electric field. The transition dipole moment measured between adjacent rotational states is consistent with the theoretical value. The coherence time and lifetime of the ground state molecules are limited by the relatively high temperature and the fact that the molecules are in an unpolarized state with unresolved hyperfine structure. Nevertheless, the coherence time is long enough to investigate both the dynamic evolution during the coherent process and the equilibrium population. Techniques such as Raman sideband cooling are expected to improve the coherence properties by allowing preparation of the atomic sample at lower temperature and in a polarized state.

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